Energy Materials Network
U.S. Department of Energy

HydroGEN
Advanced Water Splitting Materials

PEC Capability Nodes: Success Stories

Joel W. Ager, PEC Team Lead
Materials and Chemical Sciences Divisions
Lawrence Berkeley National Laboratory

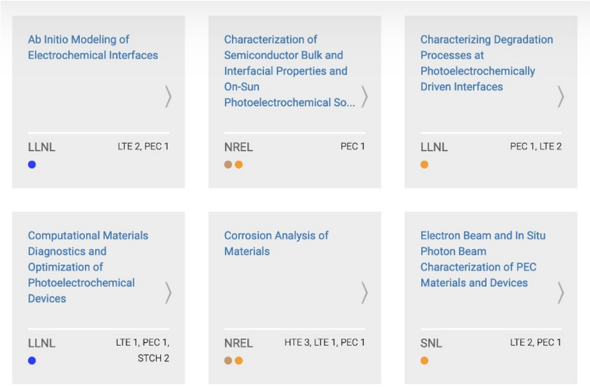
PEC Kickoff Meeting, August 22, 2023
NREL, Golden CO USA

NREL Transforming ENERGY
BERKELEY LAB Bringing Science Solutions to the World
Sandia National Laboratories
INL Idaho National Laboratory
Lawrence Livermore National Laboratory

PEC capability nodes

MENU

22 PEC 1 nodes



Ab Initio Modeling of Electrochemical Interfaces
LLNL LTE 2, PEC 1
●

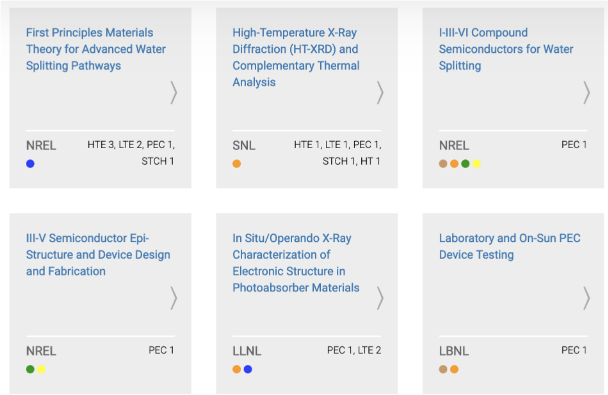
Characterization of Semiconductor Bulk and Interfacial Properties and On-Sun Photoelectrochemical So...
NREL PEC 1
●●

Characterizing Degradation Processes at Photoelectrochemically Driven Interfaces
LLNL PEC 1, LTE 2
●

Computational Materials Diagnostics and Optimization of Photoelectrochemical Devices
LLNL LTE 1, PEC 1, STCH 2
●

Corrosion Analysis of Materials
NREL HTE 3, LTE 1, PEC 1
●●

Electron Beam and In Situ Photon Beam Characterization of PEC Materials and Devices
SNL LTE 2, PEC 1
●



First Principles Materials Theory for Advanced Water Splitting Pathways
NREL HTE 3, LTE 2, PEC 1, STCH 1
●

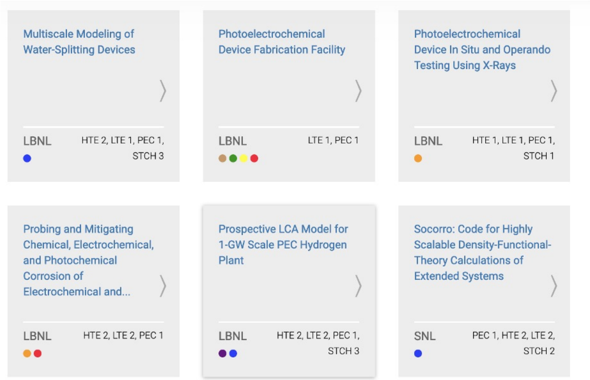
High-Temperature X-Ray Diffraction (HT-XRD) and Complementary Thermal Analysis
SNL HTE 1, LTE 1, PEC 1, STCH 1, HT 1
●

I-III-VI Compound Semiconductors for Water Splitting
NREL PEC 1
●●●●

III-V Semiconductor Epi-Structure and Device Design and Fabrication
NREL PEC 1
●●

In Situ/Operando X-Ray Characterization of Electronic Structure in Photoabsorber Materials
LLNL PEC 1, LTE 2
●●

Laboratory and On-Sun PEC Device Testing
LBNL PEC 1
●●



Multiscale Modeling of Water-Splitting Devices
LBNL HTE 2, LTE 1, PEC 1, STCH 3
●

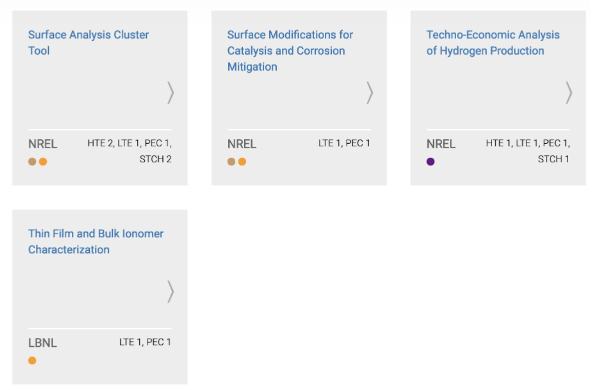
Photoelectrochemical Device Fabrication Facility
LBNL LTE 1, PEC 1
●●●●

Photoelectrochemical Device In Situ and Operando Testing Using X-Rays
LBNL HTE 1, LTE 1, PEC 1, STCH 1
●

Probing and Mitigating Chemical, Electrochemical, and Photochemical Corrosion of Electrochemical and...
LBNL HTE 2, LTE 2, PEC 1
●●

Prospective LCA Model for 1-GW Scale PEC Hydrogen Plant
LBNL HTE 2, LTE 2, PEC 1, STCH 3
●●

Socorro: Code for Highly Scalable Density-Functional-Theory Calculations of Extended Systems
SNL PEC 1, HTE 2, LTE 2, STCH 2
●



Surface Analysis Cluster Tool
NREL HTE 2, LTE 1, PEC 1, STCH 2
●●

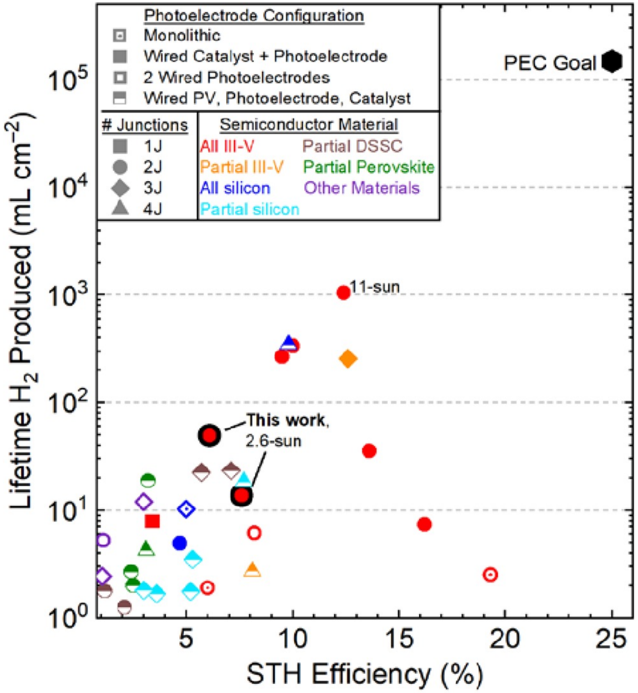
Surface Modifications for Catalysis and Corrosion Mitigation
NREL LTE 1, PEC 1
●●

Techno-Economic Analysis of Hydrogen Production
NREL HTE 1, LTE 1, PEC 1, STCH 1
●

Thin Film and Bulk Ionomer Characterization
LBNL LTE 1, PEC 1
●

HydroGEN 2.0 PEC Goal and Approach

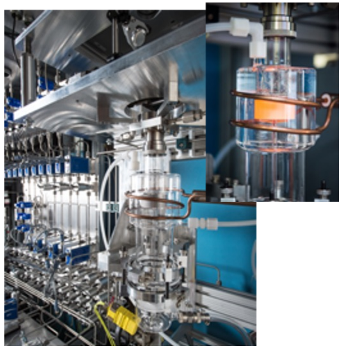
- Prioritize durability stressors and establish PEC device durability protocol
- Use density functional theory (DFT) and microkinetic modeling to describe the local environment at the electrode/electrolyte interface under operation
- Provide mechanistic understanding of PEC device degradation guided by theory and *in operando* characterization



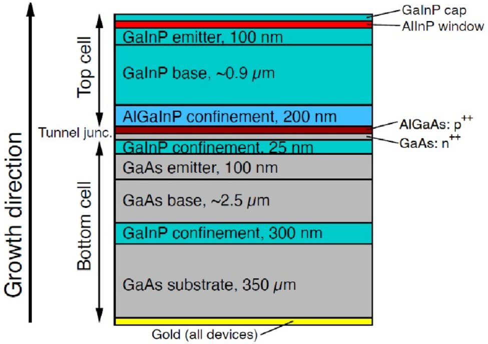
Comparison of the solar to hydrogen efficiency (STH) and lifetime H_2 produced for unassisted water splitting devices. The "PEC Goal" point in the upper right was calculated assuming a 20% capacity factor and 10-year lifetime (Ben-Naim *et al.*, *ACS Energy Lett.* **2020**, 5, 2631).

HydroGEN 2.0 PEC Status

Photoelectrode Growth by Metal Organic Vapor Phase Epitaxy

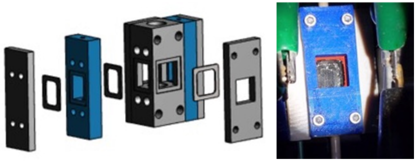


MOVPE at 620°C growth temperature



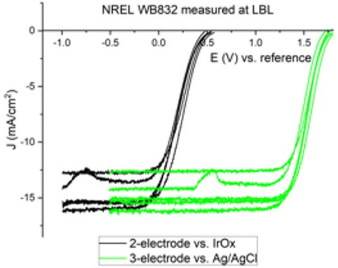
Schematic of the III-V high efficiency photoelectrode

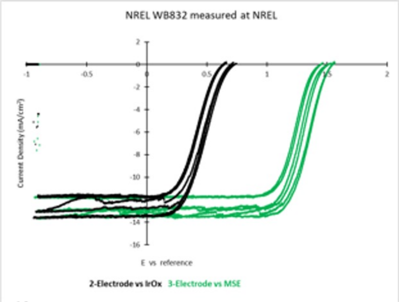
Photoelectrode Fabrication and Testing at NREL and LBNL

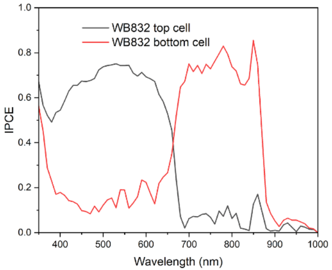


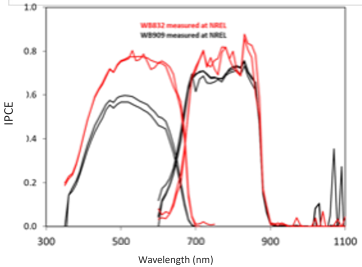
1 cm² photocathode, dual anode cell with high modularity and ease of fabrication for STH quantification

Durability and reproducibility with standard cuvette cell



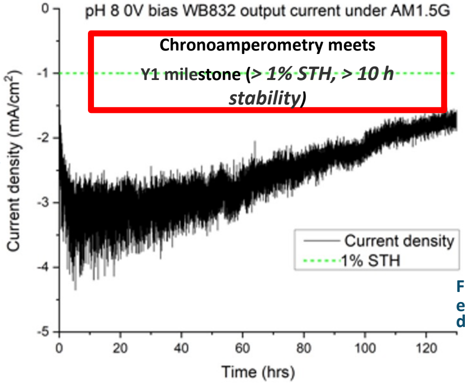






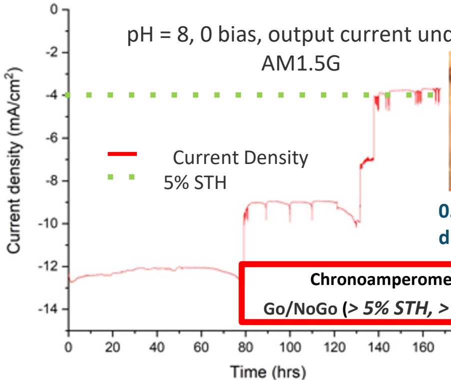
Reproducibility achieved at both Labs

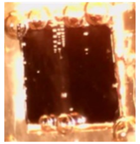
Current status



Chronoamperometry meets Y1 milestone (> *1% STH, > 10 h stability)*

Fabricated 0.2cm² electrode for durability test





0.2cm² electrode during a test

Chronoamperometry meets Go/NoGo (*> 5% STH, > 80 h stability)*

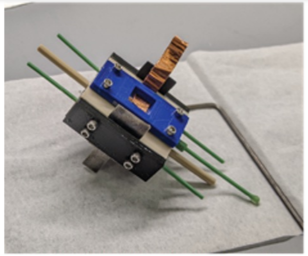
HydroGEN 2.0 Benchmarking Paper

Best Practices in PEC Water Splitting: How to Reliably Measure Solar-to-Hydrogen Efficiency of Photoelectrodes[1]

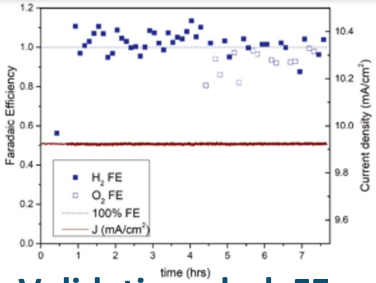
- Publication shares with PEC community best practices when measuring and reporting STH efficiency of new photoelectrodes and will improve the reproducibility of results across the field.

> *“This is a great paper for the community, thank you for dedicating the time to put this together, especially during this challenging historical time (pandemic)”*
> *–2023 attendee, Solar Fuels Gordon Conference*

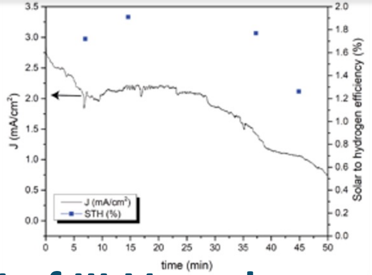
- Publication benchmarks a flow reactor for H_2/O_2 measurement, that any lab can produce from readily available materials using a CNC miller and 3D printer.
- 4098 views, 491 downloads, 10 mentions in news articles (4/10/2023)



Reactor



Validation: dark FE



STH of III-V tandem at pH 0.4

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[1] *Front. Energy Res., 27 October 2022 Volume 10 - 2022 | https://doi.org/10.3389/fenrg.2022.884364*

Selected PEC publications: Seedlings and Capability Nodes

Xiao, Y.; Kong, X.; Vanka, S.; Dong, W. J.; Zeng, G.; Ye, Z.; Sun, K.; Navid, I. A.; Zhou, B.; Toma, F. M.; Guo, H.; Mi, Z. Oxynitrides Enabled Photoelectrochemical Water Splitting with over 3,000 Hrs Stable Operation in Practical Two-Electrode Configuration. *Nat Commun* **2023**, *14*, 2047. https://doi.org/10.1038/s41467-023-37754-9.

Song, Z.; Li, C.; Chen, L.; Dolia, K.; Fu, S.; Sun, N.; Li, Y.; Wyatt, K.; Young, J. L.; Deutsch, T. G.; Yan, Y. All-Perovskite Tandem Photoelectrodes for Unassisted Solar Hydrogen Production. *ACS Energy Lett.* **2023**, *8*, 2611–2619. https://doi.org/10.1021/acsenergylett.3c00654.

Fehr, A. M. K.; Agrawal, A.; Mandani, F.; Conrad, C. L.; Jiang, Q.; Park, S. Y.; Alley, O.; Li, B.; Sidhik, S.; Metcalf, I.; Botello, C.; Young, J. L.; Even, J.; Blancon, J. C.; Deutsch, T. G.; Zhu, K.; Albrecht, S.; Toma, F. M.; Wong, M.; Mohite, A. D. Integrated Halide Perovskite Photoelectrochemical Cells with Solar-Driven Water-Splitting Efficiency of 20.8%. *Nat Commun* **2023**, *14* (1), 3797. https://doi.org/10.1038/s41467-023-39290-y.

Zeng, G.; Pham, T. A.; Vanka, S.; Liu, G.; Song, C.; Cooper, J. K.; Mi, Z.; Ogitsu, T.; Toma, F. M. Development of a Photoelectrochemically Self-Improving Si/GaN Photocathode for Efficient and Durable H_2 Production. *Nat. Mater.* **2021**, *20*), 1130–1135. https://doi.org/10.1038/s41563-021-00965-w.

Gaillard, N.; Septina, W.; Varley, J.; Ogitsu, T.; Ohtaki, K. K.; Ishii, H. A.; Bradley, J. P.; Muzzillo, C.; Zhu, K.; Babbe, F.; Cooper, J. Performance and Limits of 2.0 EV Bandgap $CuInGaS_2$ Solar Absorber Integrated with CdS Buffer on F:SnO_2 Substrate for Multijunction Photovoltaic and Photoelectrochemical Water Splitting Devices. *Mater. Adv.* **2021**, *2*, 5752–5763. https://doi.org/10.1039/D1MA00570G.

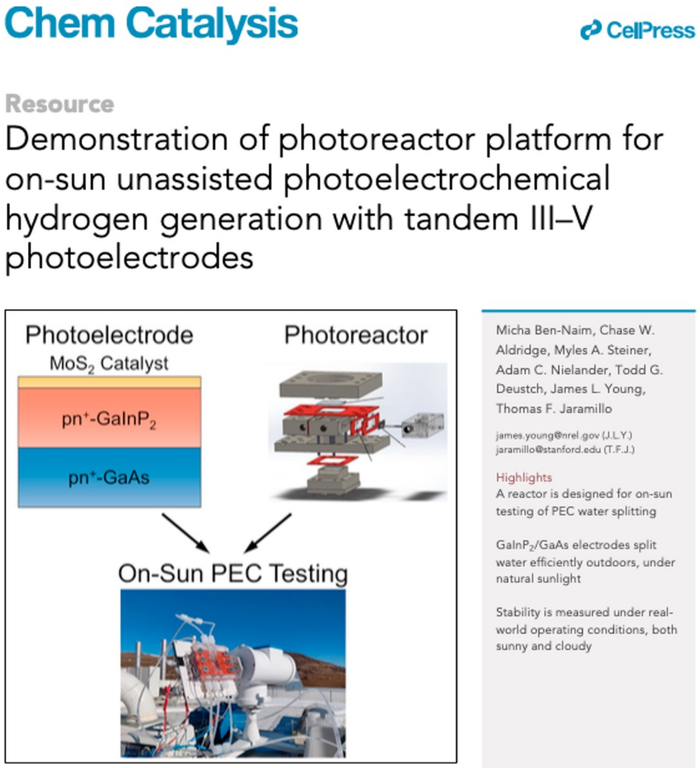
SPRINGER NATURE

Behind the Paper

The real stories behind the latest research papers, from conception to publication, the highs and the lows

NREL collaboration with Jaramillo (Stanford)

NREL nodes provided III-V samples and hosted two Stanford graduate students for training on outdoor photoreactor measurements



Chem Catalysis CellPress

Resource

Demonstration of photoreactor platform for on-sun unassisted photoelectrochemical hydrogen generation with tandem III–V photoelectrodes

Micha Ben-Naim, Chase W. Aldridge, Myles A. Steiner, Adam C. Nielander, Todd G. Deustch, James L. Young, Thomas F. Jaramillo

james.young@nrel.gov (J.L.Y.)
jaramillo@stanford.edu (T.F.J.)

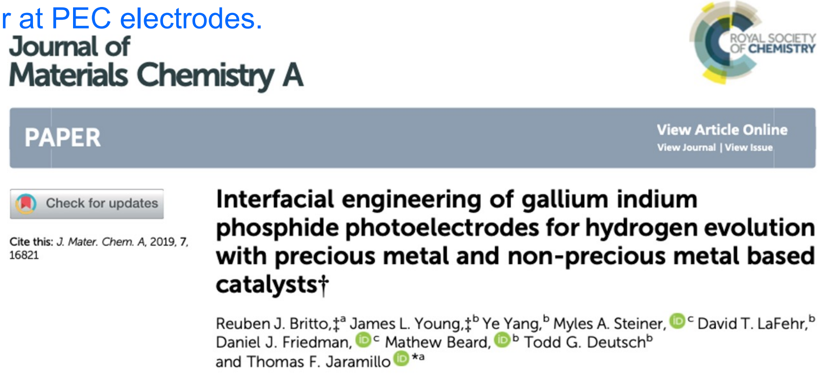
Highlights

A reactor is designed for on-sun testing of PEC water splitting

$GaInP_2$/GaAs electrodes split water efficiently outdoors, under natural sunlight

Stability is measured under real-world operating conditions, both sunny and cloudy

NREL nodes provided III-V samples and cocatalysts surface modifications. Node experts also connected with another NREL group to perform transient photoreflectance spectroscopy to understand interfacial charge transfer at PEC electrodes.



Journal of Materials Chemistry A

ROYAL SOCIETY OF CHEMISTRY

PAPER

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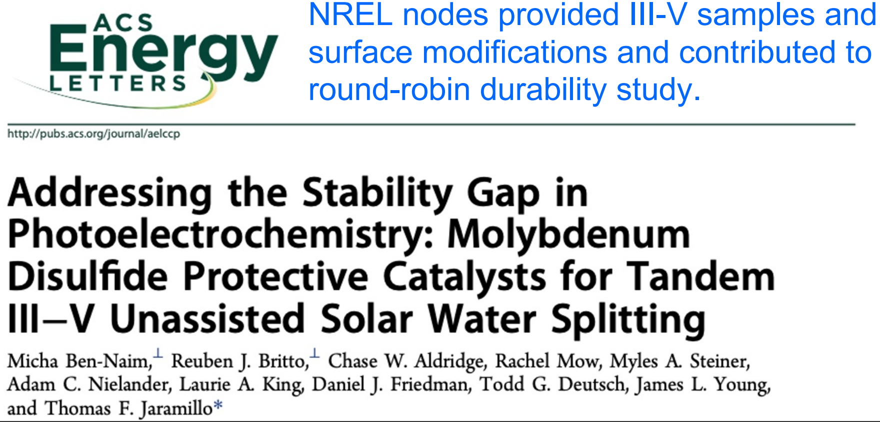
Cite this: J. Mater. Chem. A, 2019, 7, 16821

Interfacial engineering of gallium indium phosphide photoelectrodes for hydrogen evolution with precious metal and non-precious metal based catalysts†

Reuben J. Britto,‡[a] James L. Young,‡[b] Ye Yang,[b] Myles A. Steiner,[c] David T. LaFehr,[b] Daniel J. Friedman,[c] Mathew Beard,[b] Todd G. Deutsch[b] and Thomas F. Jaramillo*[a]

NREL nodes provided III-V samples and surface modifications and contributed to round-robin durability study.



ACS Energy LETTERS

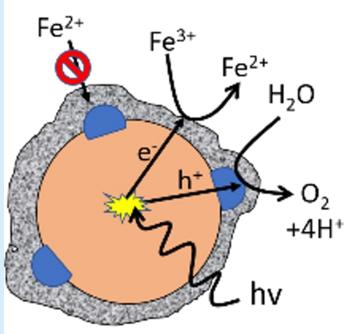
http://pubs.acs.org/journal/aelccp

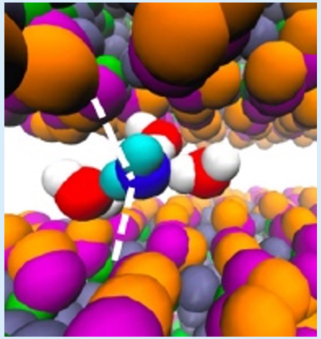
Addressing the Stability Gap in Photoelectrochemistry: Molybdenum Disulfide Protective Catalysts for Tandem III–V Unassisted Solar Water Splitting

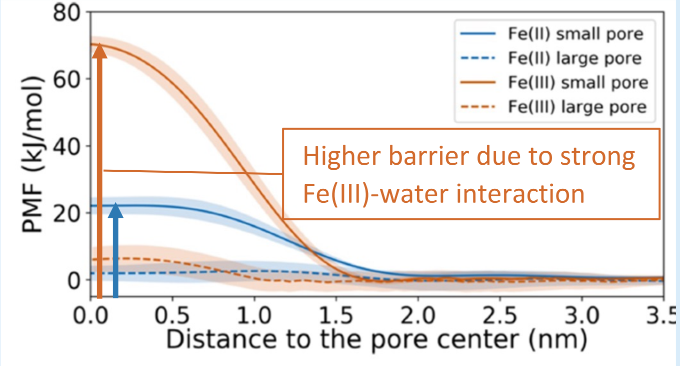
Micha Ben-Naim,[⊥] Reuben J. Britto,[⊥] Chase W. Aldridge, Rachel Mow, Myles A. Steiner, Adam C. Nielander, Laurie A. King, Daniel J. Friedman, Todd G. Deutsch, James L. Young, and Thomas F. Jaramillo*

LLNL collaboration w UCI/LLNL-LBNL collaboration w U. Hawaii

UCI: size of nano-pore & selective permeation



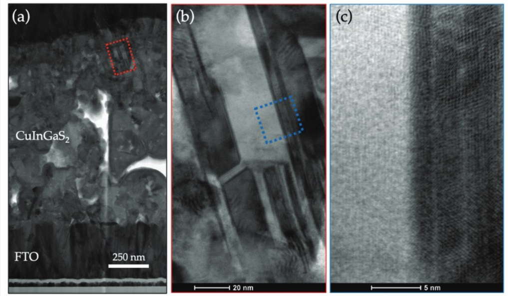


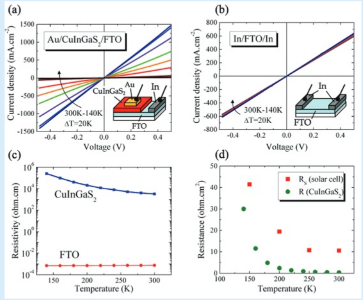


Higher barrier due to strong Fe(III)-water interaction

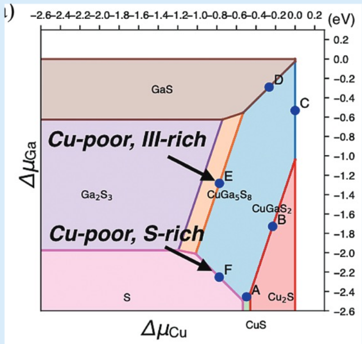
UCI seedling, ACS Appl. Mater. Interfaces, doi:10.11021/acsami.2c22865

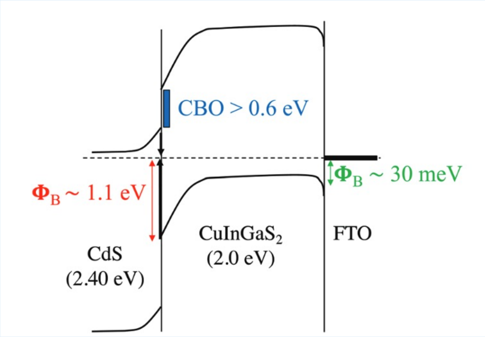
Hawaii: band alignment of $CuInGaS_2$/CdS/SnO_2





Alloy phase diagram and possible band alignments





Gaillard et al. Mater. Adv. **2**, 5752 (2022)

LLNL-LBNL collaboration with U. Michigan on elucidating the origin of GaN performance

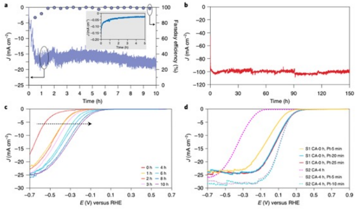


Fig. 1 | Self-improving behaviour of Si/GaN photocathodes. a, CA testing for 10 h under 1-sun illumination and constant bias at −0.6 V versus RHE, the corresponding Faradaic efficiency reveals the self-improving nature of GaN. Inset: CA testing on bare Si for 5 h under 1-sun illumination and −0.6 V versus RHE, this bare Si photocathode rapidly drops down to <0.05 mA cm^{-2} within an hour. **b**, Accelerated CA testing on bare epilayer Si/GaN photocathode at constant bias of −0.6 V versus RHE under 3.5-sun illumination for 150 h in 0.5 M H_2SO_4. **c**, Photocurrent density versus applied electrochemical potential (*J*–*E*) curves as a function of CA testing time. **d**, Several selected *J*–*E* curves of as-prepared Si/GaN photocathode (CA-0 h) and 4 h CA-tested Si/GaN photocathode (CA-4 h) with different Pt photodeposition durations under 1-sun illumination.

Improve hydrogen evolution reaction (HER) efficiency observed at the initial stage of experiment

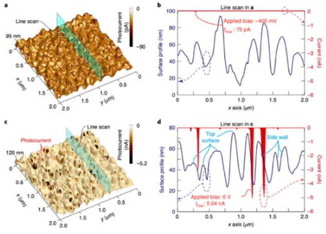


Fig. 2 | Photoconductive AFM characterization on CA-0 h and CA-10 h samples. a,c, The topography of the CA-0 h sample (**a**) and CA-10 h sample (**c**). **b,d**, Corresponding surface profile and photocurrent extracted from the line scans for the CA-0 h sample (**b**) and CA-10 h sample (**d**).

The improvement of photocurrent is localized at the sidewalls, likely (1010) surface

Nature Materials **20**, 1130 (2021)

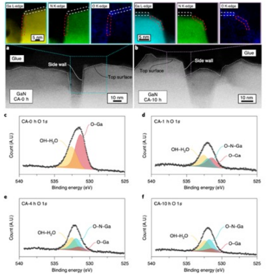
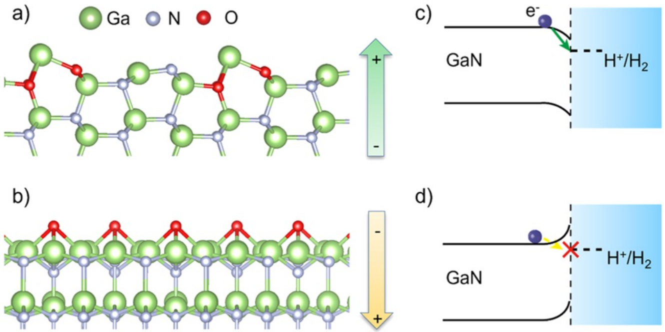


Fig. 3 | Chemical analysis of Si/GaN photocathode surface. a, STEM image of CA-0 h surface with insets (left to right) showing the EELS mappings of Ga L-edge, N K-edge and O K-edge. **b**, STEM image of CA-10 h surface with insets (left to right) showing the EELS mappings of Ga L-edge, N K-edge and O K-edge. **c–f**, O 1s core-level spectra of CA-0 h Si/GaN photocathode (**c**); Si/GaN photocathode after 1 h CA testing (**d**); Si/GaN photocathode after 4 h CA testing (**e**); and Si/GaN photocathode after 10 h CA testing (**f**).

The sidewall surface develops a composition similar to gallium oxynitride



DFT simulations show that alternating substitution of N with O at the first two layers of GaN(1010) surface provides the band-bending that facilitate charge transfer at the interface, which is consistent with improved HER activity of GaN(1010)

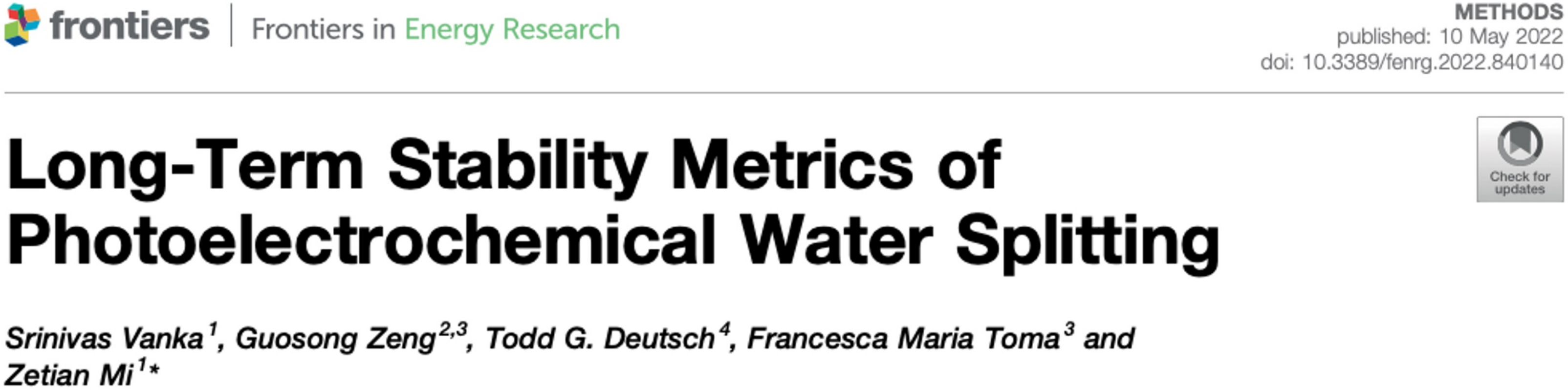
U Michigan seedling project on GaN supported by PEC capability nodes at LLNL, Nano Lett. **22**, 2236 (2022)

Joint LBNL and NREL collaborations with seedlings

LBNL and NREL nodes advised Rice on photoreactor design and water oxidation catalysts. LBNL contributed to FE measurements and sent a photoreactor to Rice.

"Integrated halide perovskite photoelectrochemical cells with solar-driven water-splitting efficiency of 20.8%" Austin M. K. Fehr, Ayush Agrawal, Faiz Mandani, Christian L. Conrad, Qi Jiang, So Yeon Park, Olivia Alley, Bor Li, Siraj Sidhik, Isaac Metcalf, Christopher Botello, James L. Young, Jacky Even, Jean Christophe Blancon, Todd G. Deutsch, Kai Zhu, Steve Albrecht, Francesca M. Toma, Michael Wong & Aditya D. Mohite. *Nature Communications* volume 14, Article number: 3797 (2023).

LBNL node performed durability measurements and advised seedling on testing protocol. NREL and LBNL nodes contributed to manuscript draft and revisions



frontiers | Frontiers in Energy Research

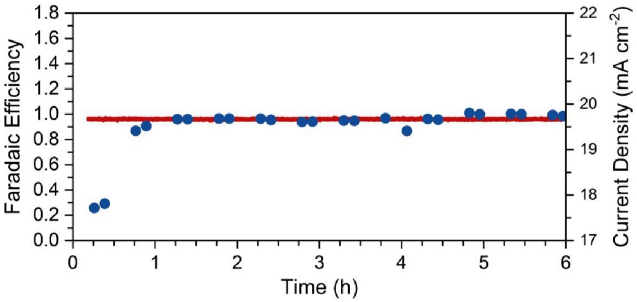
METHODS
published: 10 May 2022
doi: 10.3389/fenrg.2022.840140

Long-Term Stability Metrics of Photoelectrochemical Water Splitting

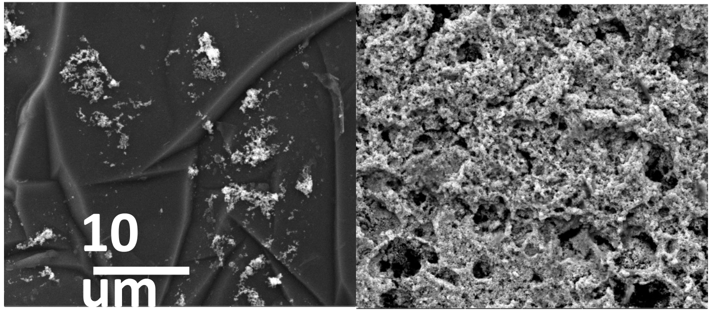
Srinivas Vanka[1], Guosong Zeng[2,3], Todd G. Deutsch[4], Francesca Maria Toma[3] and Zetian Mi[1]*

Specific supports of Rice seedling by LBNL

- PEC device fabrication facility for design of cell to measure FE and STH (HydroGEN 2.0 cell developed in the supernode), and also for early sputter deposition of catalysts
- Laboratory PEC device testing for measurements of FE and STH prior to materials and expertise transfer of these methods to Rice
- SEM, XPS, ICPMS measurements for probing and mitigating corrosion
- Lead author of Nature Communications paper visited LBNL to continue learning cell design and perform measurements at LBNL’s facility



Pt/C catalyst measured at LBNL to have an average Faradaic efficiency to H_2 of 98% +/- 2%.



Delaminated vs intact regions post-run

SEM at LBNL showed widespread delamination of an early catalyst/CAB post-testing

Awards at FY2021 HFTO AMR

Hydrogen Technologies—Production
Aditya Mohite, William Marsh Rice University

This award is presented to Dr. Aditya Mohite of Rice University for achieving an integrated halide perovskite photoelectrochemical cell with record-setting solar-driven water-splitting efficiency of 20.8%. Mohite's system exhibited extended durability due to a novel graphite anticorrosion barrier that translates >99% of photoelectric power directly to hydrogen production. His work increasing the durability and efficiency of photoelectrochemical systems has led the way toward deployment of this renewable hydrogen production technology at scale.

https://www.hydrogen.energy.gov/annual-review/annual_review22_awards.html

Impact on field

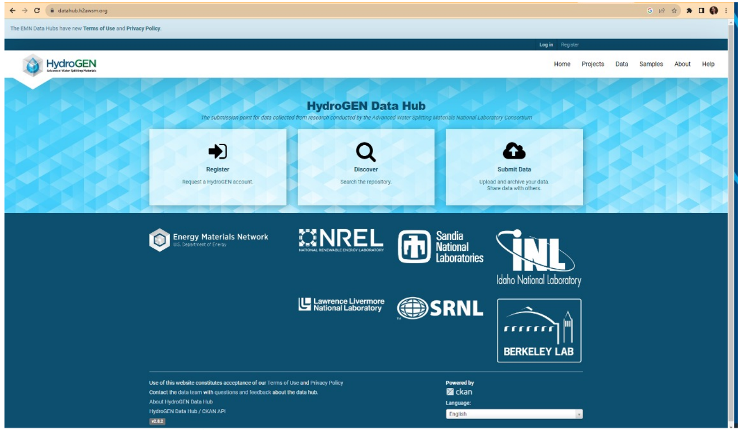
The HydroGEN EMN data team led by NREL and LBNL worked together to make the first full database of all the data included in the Nature Materials paper with a related DOI

doi.org/10.17025/1764132, doi.org/10.17025/1764126, doi.org/10.17025/1764127

doi.org/10.17025/1764161, doi.org/10.17025/1764162, doi.org/10.17025/1764163

doi.org/10.17025/1764157, doi.org/10.17025/1764159, doi.org/10.17025/1764133

doi.org/10.17025/1764156, doi.org/10.17025/1764155, doi.org/10.17025/1764154



datahub.h2awsm.org

The EMN Data Hubs have new **Terms of Use** and **Privacy Policy**.

HydroGEN Data Hub

The submission point for data collected from research conducted by the Advanced Water Splitting Materials National Laboratory Consortium

Register
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Search the repository.

Submit Data
Upload and archive your data.
Share data with others.

Use of this website constitutes acceptance of our Terms of Use and Privacy Policy

Contact the data team with questions and feedback about the data hub.

About HydroGEN Data Hub

HydroGEN Data Hub / CKAN API

v2.8.2

Powered by ckan

Language: English

Summary – impact of capability nodes “beyond the paper”

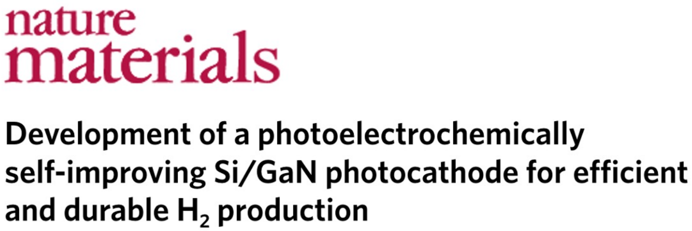
ACS Energy Letters

http://pubs.acs.org/journal/aelccp

All-Perovskite Tandem Photoelectrodes for Unassisted Solar Hydrogen Production

nature communications

Oxynitrides enabled photoelectrochemical water splitting with over 3,000 hrs stable operation in practical two-electrode configuration



nature materials

Development of a photoelectrochemically self-improving Si/GaN photocathode for efficient and durable H_2 production

“My exposure to LBNL through seedlings opened the path for me to do further research on-site through the DOE SCGSR program, which was transformative for my career.”
--Austin Fehr

“…a pure mechanical engineering background person [such] as me who can work in the HydroGEN project and win the The Hydrogen and Fuel Cells Postdoctoral Recognition Award. This experience itself already explains how amazing this HydroGEN/LBL is, i.e., providing fair opportunities to all people and delivering the best research environment for them to succeed in science and technology.
--Guosong Zeng

“HydroGEN has uniquely enabled my professional development in many dimensions. It allowed me to expand my knowledge from basic to applied science, to establish new collaborations with the top scientists at different National Labs and in Academia, and to grow as a scientific leader.”
--Francesca Toma